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Novel Electroceramic Materials and Integrated Devices

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DAAD19-99-1-0362

6. AUTHOR(S)
Ram S. Katiyar

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University of Puerto Rico - San Juan

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10. SPONSORING / MONITORING AGENCY REPORT NUMBER
ARO 39690.12-PH-SAH

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13. ABSTRACT (Maximum 200 words)

A number of Perovskites have been grown and their properities analyzed under the general field of "Novel Electroceramic Materials and Integrated Devices". Besides their structural properties, their electrical and optical properties were investigated. These investigations were published in ten papers that have appeared in scientific journals.

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Technical Report (Final)

Grant: DAAD 19-99-1-0362 ARO Proposal Number: P-39690-PH-SAH

Performance Period: September 1, 1999 - August 31, 2000

PI: Dr. Ram S. Katiyar

SUMMARY

Lead Based Perovskites:

Lead based PLT, PLT:Ce, PLT:Gd, PZT, and PZT:Nd materials were synthesized using sol-gel and rf sputtering techniques and the effect of film thickness, dopants and precursor solvents was studied in them.

Thickness Effect in PZT: With Zr substitution on the B sites of PT lattice the soft mode intensity approached zero near the morphotropical phase boundary. In PZT (53/47) thin films of different thicknesses (100, 200, 369, 492, and 755 nm) on Pt, the soft mode was observed at about 68 cm^{-1} in the 100 nm thick film. This frequency decreased to 57 cm^{-1} with increasing thickness up to 755 nm. But, no soft mode was detected in the Raman spectrum of powders prepared from the same solution, which is supposed to be stress free. The presence of soft mode in films was therefore, attributed to the presence of strain in films. The two dimensional stress in these films causes the soft mode to appear. These stresses were found considerably reduced in powders and fibers prepared under the similar conditions. A comparative study of PT/PLT films and fibers, prepared by sol-gel technique, suggested less stresses in the fibers. The variation of stress was correlated to the micro structural features in these structures.

La Modified PT (PLT): The phase transition temperature and relaxor behavior was studied in $Pb_{1-x}La_xTi_{1-x/4}O_3$ thin films using temperature dependent Raman scattering. The x-ray diffraction measurements indicated a cubic structure for $x>0.15$, while the Raman modes were observed up to $x=0.25$. From thermal studies it was found that the organic removal and crystallization starts at about 315 and 500C, respectively. AFM estimated surface roughness was found to be decreasing with the La doping. Raman scattering, in the temperature range 75-783K, suggested the transition temperature about 730, 680, 614, and 563 K for $x=0.05$, 0.10, 0.15, and 0.20 compositions, respectively. The diffuse nature of the FE phase transition owing to a short-range structural disorder in the paraelectric cubic phase and an increase in the diffuseness with La doping indicated the relaxor behavior and its relationship with the local disorder in these materials.

The substrate and precursor effects in sol-gel derived $Pb_{0.9}La_{0.15}TiO_3$ thin films were studied at various annealing temperatures in the range of 350 – 650 ^{0}C. X-ray results indicated that films prepared by acetic acid route had texturing along (100) orientation that increased with increasing annealing temperature. These films also exhibited better FE properties on Pt compared to Pt/Si substrates.

Rare Earth (Ce and Gd) Doped PLT: Isovalent substitution of rare earths (Ce^{+3} and Gd^{+3}) on the La^{+3} sites of the sol-gel prepared $Pb_{0.85}La_{0.15}TiO_3$ films was investigated. With increasing contents of Gd, a decrease in lattice parameters, and increase in the tetragonality ratio (*c/a*) was observed. Relatively less FE ordering was obtained in Ce doped thin films. Raman spectra exhibited features characteristics of bulk $PbTiO_3$, including the observation of the soft mode. Variation of the soft phonon mode in these films was investigated as a function of the composition *x*, (in the range 0.00 to 0.15) and temperature (in the range (25-600^{0}C). Unlike the rare earth doping in PT, the isovalent substitution in PLT was found to increase the

transition temperature with increasing Gd and Ce contents. In Ce doped films the T_c was found to be lower for x =0.07 film compared to x =0.05. The precipitation of Ce in 7 at% Ce doped films suggests lower solubility limit of Ce (about 5at%) compared to that of Gd. A discontinuity in the polarization and dielectric values was also observed at x =0.05 composition of Ce doped films.

<u>Rare Earth (Nd, Gd, and Ce) Doped PZT(53/47)</u>: We have studied the effect of rare earth dopants on the phase formation behavior and electrical properties of sol-gel derived PZT (%3/47) thin films. In all these films the perovskite phase was obtained up to 5 at% doping and beyond that pyrochlore phase was formed to coexist with the perovskite phase. Ce and Gd dopings (1 - 2 at%) exhibited improved FE and dielectric properties as compared to the undoped PZT films. Nd doping (2 at%) was found to be effective to increase the retained switchable polarization of undoped PZT from 63 to 84 %. The transition temperature of undoped PZT was found to be reduced with Nd doping. The Nd doped films also exhibited typical relaxor behavior with a diffuse phase transition. This may be due to disorder in the B-site of the perovskite lattice caused by the Nd doping.

Layered Perovskites:

Layered structures of $Ba_xSr_{1-x}TiO_3$, $Bi_4Ti_3O_{12}$, $Bi_{4-x}La_xTi_3O_{12}$, $Sr_{1-x}Ba_xBi_2Ta_2O_9$, $Sr_{1-x}Ba_xBi_2TaNbO_9$, $SrBi_2Ta_{2-x}Nb_xO_9$, {1-x$SrBi_2Ta_2O_9$ - xBi_3TiTaO_9}, {1-x$SrBi_2Nb_2O_9$ - xBi_3TiNbO_9} for x = 0 to 1, were synthesized. All materials were prepared in the powders and thin film forms by spin coating. Thin films of FE $(SrBi_2Ta_2O_9)_x$(Bi3TiNbO9)$_{1-x}$ layered structure were prepared for x = 0.0, 0.2, 0.4, 0.6, 0.8, and 1.0, by metal organic solution deposition method on Pt/TiO_2/SiO_2/Si substrates. The Raman spectrum for the film with x = 0 shows bands around 60, 170, 232, 256, 337, 569, and 839 cm^{-1}, which indicate Bi_3TiNbO_9 (BTN) formation. The prominent band around 839 cm^{-1}, which is an A_{1g} mode of the orthorombic symmetry, corresponds to symmetric stretching of the BO_6 octahedra. The frequency of this band is found to shift significantly as $SrBi_2Ta_2O_9$ (SBT) material is added to the BTN compound. The Raman spectra show frequency shifts and a broadening of the bands as x change from 0 to 0.4, which is related to the differences in mass between Sr and Bi in the A-sites, and Ta, Ti, and Nb in the B-sites. A comparison between film and bulk indicates that the crystallization degree in the film is lower than that in the bulk, which can be due to the presence of stress in the films. The temperature-dependent study of the films indicates a strong contribution from defects in the Raman spectra.

Thin films of $SrBi_2(Ta_xNb_{1-x})O_9$ with x = 0.8 and x = 2.0 were also grown on Si(100), MgO(100), Pt/TiO_2/SiO_2/Si substrates using PLD technique. The influence of growth conditions such as substrate temperature, oxygen partial pressure, etc. on the structural properties of the films were analyzed. In the films prepared at substrate temperatures between 25 °C to 750 °C, it was observed that only the films deposited above 600°C substrate temperature crystallize into the desired phase. There is no significant influence on oxygen partial pressure (100-450 mTorr) on the crystallization behavior of these films deposited at various substrate temperature. X-ray photoelectron spectroscopy studies of Sr 3d level for the films with composition $SrBi_2Ta_{0.8}Nb_{1.2}O_9$ suggest the oxygen ions in the $Sr(TaNb)_2O_7$ perovskite layers to be much more stable than those in Bi_2O_2 layers. Micro-Raman study of $SrBi_2Ta_{.2}O_9$ films deposited on Pt/TiO_2/SiO_2/Si show the presence of FE phase. The $SrBi_2Ta_{.2}O_9$ films deposited at 700°C with 100 mTorr oxygen pressure and 1.3 J/cm^2 laser fluence resulted in 8.9 $\mu C/cm^2$ spontaneous polarization, 4.42 $\mu C/cm^2$ remnant polarization and 39.4 KV/cm coercive field values.

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RÍO PIEDRAS CAMPUS
FACULTY OF NATURAL SCIENCES

DEPARTMENT OF PHYSICS

May 9, 2001

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Reference: **A.R.O. Proposal: P-39690-PH-SAH Grant #DAAD19-99-1-0362**

Dear Ms. Ashe:

Please find enclosed a copy of the documents (accounting and technical reports) that were submitted to your office on November 30, 2000 and February 21, 2001 respectively. On the advice of Dr. Mikael Ciftan (Program director) we are herewith submitting the enclosed copies as well as a summary of the technical report. Please acknowledge the receipt. Thanking you.

Sincerely yours,

R S Katiyar

Prof. Ram S. Katiyar
Director of SPECLAB
Advanced Materials Research Group
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P-39690-PH-SAH

February 21, 2001

Mrs. Sandra Yates / U.S. Army Research Office
4300 South Miami Boulevard
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Dear Mrs. Yates:

Enclosed you will find the report of grant number DAAD1999-1-0362. The title of our research grant is "Novel Electroceramic Materials and Integrated Devices." I have now prepared a report and I am sending it to you via Federal Express.

We look forward to continue our mutually beneficial relationship. Please let me know if you need further details about the report. Our telephone number is 787-751-4210 or via fax at 787-764-2571.

Sincerely,

R S Katiyar

Prof. Ram S. Katiyar
Director of SPECLAB
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SPECLAB

Advanced Materials Research Group

List of publications of research made possible by the following Grant: DAAD 19-99-1-0362
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Publications

1. Growth, microstructure and micro-Raman studies of RF magnetron sputter deposited $SrBi_2Ta_2O_9$ and $SrBi_2TaNbO_9$ films, Menka Jain, Srinivas Sathiraju, and Ram S. Katiyar, *Proceedings of Materials Research Society,* **580**, 345 (2000).

2. Raman spectroscopy study of phase transitions in self-assembled nanostructures: (1-x)PZN-xPT, S. Gupta, R. S. Katiyar, and A. S. Bhalla, *Proceedings of The XVIIth International Conference on Raman Spectroscopy*, (Editors S.-L. Zhang and B.-F. Zhu, John Wiley & Sons) pp. 448, (2000).

3. Light scattering from pulsed laser deposited $BaBi_4Ti_4O_{15}$ thin films, R.K. Soni, Anju Dixit, R.S. Katiyar, A. Pignolet, K.M. Satyalakshmi and D. Hesse, *Materials Research Society Proceedings,* **623**, 167 (2000).

4. Structural and electrical properties of $Sr_{1-x}Ba_xBi_2Ta_2O_9$ thin films, R. Melgarejo, M.S. Tomar, P.S. Dobal, and R.S. Katiyar, *Thin Solid Films*, (Accepted).

5. Synthesis of $Zn_{1-x}Mg_xO$ and its structural characterization, M.S. Tomar, R.E. Melgarejo, P.S. Dobal, and R.S. Katiyar, *Journal of Materials Research,* (Submitted).

6. Growth and properties of $Sr_{1-x}Ba_xBi_2TaNbO_9$ materials and thin films, M.S. Tomar, R.E. Melgarejo, P.S. Dobal, M. Jain, R.S. Katiyar, *Journal of Materials Science,* (Submitted).

7. {1-x $SrBi_2Ta_2O_9$-xBi_3TiTaO_9} materials: structural behavior and ferroelectric response, R.E. Melgarejo, M.S. Tomar, P.S. Dobal, S.K. Filippov, R.S. Katiyar, and K.A. Kuenhold, *Journal of Materials Science and Engineering (B),* (Submitted).

8. Effect of rare earth doping on Lead based Perovskite thin films, S.B. Majumder, P.S. Dobal, B. Roy, S. Bhaskar, and R.S. Katiyar, *I.E.E.E. Symposium Proceedings* (Submitted).

9. Effect of rare earth doping on Sol-gel derived PZT thin films, S.B. Majumder, P.S. Dobal, B. Roy, S. Bhaskar, and R.S. Katiyar, *Ferroelectric Letters*, (Submitted).

10. Structural studies of $Bi_{4-x}M_xTi_3O_{12}$ (M = La, Nd) ferroelectric materials, M.S. Tomar, R.E. Melgarejo, P.S. Dobal, A. Dixit, and R.S. Katiyar, *Journal of Applied Physics* (Submitted).

Scientific Presentations

1. Growth, microstructure and Micro-Raman studies of RF magnetron sputter deposited $SrBi_2Ta_2O_9$ and $SrBi_2TaNbO_9$ films, Menka Jain, Srinivas Sathiraju, and Ram S. Katiyar, *Materials Research Society Fall Meeting*, Boston, Nov.29 - Dec 3, 1999.

2. Micro-Raman study of self -assembled nanostructures: (1-x)PZN : xPT solid solution, S. Gupta, R. S. Katiyar, R. Guo, and A. S. Bhalla, *Materials Research Society Fall Meeting*, Boston, Nov.29 - Dec 3, 1999.

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Spectroscopy Laboratory

List of publications from projects made possible by the following Grant: DAAD 19-99-1-0362
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1. Growth, microstructure and Micro-Raman studies of RF magnetron sputter deposited $SrBi_2Ta_2O_9$ and $SrBi_2TaNbO_9$ films, Menka Jain, Srinivas Sathiraju, and R. S. Katiyar, *Proceedings of Materials Research Society*`, **580**, (1999).

2. Micro-Raman study of self-assembled nanostructures: (1-x)PZN : xPT solid solution, S. Gupta, R. Guo, A. S. Bhalla, and R. S. Katiyar, *Proceedings of Materials Research Society,* **581**, (1999).

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Sincerely,

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